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On the Changes of Viscosity Preceding the Precipitation of Aluminium Hydrate from Aluminate Solutions

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In order to ascertain whether and in what way the structure of clear aluminate solutions is modified prior to the precipitation of aluminium hydrate, the viscosity and specific gravity of these solutions were measured from the moment of their preparation to the moment of incipient precipitation.

The experiments were performed both on pure aluminate solutions and on solutions to which methanol had been added. The solutions contained from 0.916 to 1.029 *M* Al_2O_3/1. and from 1.427 to 1.567 *M* Na_2O/1., the molal ratio Al_2O_3/Na_2O varied from 1: 1.445 to 1:1.556.

It was found that the viscosity of both the pure aluminate solutions and the solutions with the addition of methanol rose and fell periodically until a maximum was reached. At this moment precipitation of aluminium hydrate set in, and the viscosity dropped suddenly. A tentative theoretical explanation of these phenomena is proposed.

It was found that equal amounts of methanol caused a larger increase in viscosity of sodium hydroxide and aluminate solutions than of water. Since equal amounts of methanol caused the same increase in viscosity of sodium hydroxide solutions and of aluminate solutions, the conclusion is warranted that in both those solutions the same chemical processes take place.

In sodium hydroxide solutions of different concentrations methanol combines with the hydroxide to form a molecular combination having a molal ratio methanol : NaOH = 1 : 1. The same conclusion applies to aluminate solutions.

It is shown by a simple calculation that in aluminate solutions to which methanol has been added, polyaluminates are present in considerable concentrations. Assuming that only one kind of aluminate ions is present, it is probable that this is the trialuminate hydroxo-complex ion $Al_3(OH)_{10}^-$.

Different opinions have been advanced on the structure of aluminate solutions: according to some authors (Fricke[1], Dhar and Ghosh[2], Mohanlal and Dhar[3]) they are suspensions of aluminium hydrate in sodium hydroxide solutions, the hydrate being in a fine dispersion comparable to a colloidal solution; Heyrowsky[4], Jahr and Plaetschke[5], Bode[6], Brosset, Biedermann and Sillén[7] consider them to be true molecular solutions of aluminates, mainly meta-aluminates.

Herrmann[8] presumes that diluted aluminate lyes represent solutions in which the sodium hydroxide acts as solvent and in which weak aluminate complexes between aluminium hydrate and hydroxyl ions are formed.

Our own investigations aim at contributing to the elucidation of these problems, which are not only of interest from a theoretical point of view, but may also lead to a better understanding and, consequently, to a more efficient operation of the Bayer process for the manufacturing of alumina.

The starting-point of our investigations is the assumption that the structure of the metastable aluminate solutions must necessarily undergo some sort of modification before the precipitation of aluminium hydrate $Al_2O_3 \cdot 3H_2O$ sets in. Evidence brought forward by Heyrowsky[9] (preparation of solid lithium aluminate from an aluminate solution) points to the conclusion that aluminate solutions, at least when diluted, are really what their name implies, i. e. solutions of sodium aluminate $NaAl(OH)_4$ resp. $NaAl_2(OH)_7$. In the face of such evidence the hypothesis of aluminate solutions as some sort of crypto-colloidal solutions of aluminium hydrate in sodium hydroxide solution, and of precipitation of gibbsite or bayerite from this solution as being comparable the crystallisation of a salt from a supersaturated solution, appears to be untenable. However, the assumption that the solutions contain solely the above mentioned sodium aluminates is also unsatisfactory, since the electrical conductivity of the solution does not increase in proportion with the quantity of hydrate precipitated as it should do if the decomposition of the aluminate ions took place according to the equations:

$$\begin{aligned} Al(OH)_4^- &\longrightarrow Al(OH)_3 + OH^- \quad \text{or} \\ 2\,Al(OH)_4^- &\longrightarrow Al_2O_3 \cdot 3H_2O + 2OH^- \quad \text{or} \quad . \; . \; . \; . \; . \; . \; . \; . \\ Al_2(OH)_7^- &\longrightarrow Al(OH)_3 + Al(OH)_4^- \longrightarrow 2Al(OH)_3 + OH^- \end{aligned} \tag{1}$$

In order to ascertain whether and in what way the structure of aluminate solutions is modified by ageing, we measured the viscosity and specific gravity of such solutions from the moment of their preparation to the moment of incipient precipitation. To make the effect more readily measurable, rather concentrated solutions were used. On the other hand, the rate of structural changes had to be reduced in order to make it measurable, but not so much as to unduly increase the duration of the experiments. As it is well known metastable solutions possesing a caustic modul higher than 1.8 may be allowed to stand for months under favorable circumstances without exhibiting any change. Assuming that the process in the solution is a veritable chemical change, all the experiments were carried out at a relatively low temperature of $1 \pm 0.1^{0}C$ in order to obtain larger differences of viscosity, to reduce the reaction rate and to shift the equilibrium towards the precipitation of aluminium hydrate.

For the same reasons aluminate solutions having a low caustic ratio ($\alpha = M\ Na_2O/M\ Al_2O_3$) were employed (from 1.445 to 1.556). The solutions contained from 0.916 to 1.029 M Al_2O_3/l. and from 1.427 to 1.567 M Na_2O/l.

It must be pointed out that it is very difficult to prepare a number of samples of aluminate solutions having exactly the same composition, being impossible to prevent the loss of small quantities of solution when dissolving metallic aluminium. Moreover, the properties of aluminate solutions are not functions of the composition only, but depend also on some fortuitous circumstances in their preparation: two aluminate solutions prepared under the same circumstances as far as we can control them, may not be quite identical in their properties. They may show, for instance, differences in their viscosity or electrical conductivity. We were, therefore, obliged to compare the properties of aluminate solutions having only approximately the same composition and to verify our results by replicating our experiments.

EXPERIMENTAL

Preparation of aluminate solutions

About 100 g. of sodium hydroxide (p. a.) were dissolved in approximately 300 ml. of distilled water previously freed of carbon dioxide by boiling, and placed in a nickel vessel holding 2 liters. 42.0 g. of granulated pure aluminium was added to the warm solution in small portions. The dissolution of the aluminium was generally completed in about 20 minutes. The vessel was covered with a watch glass, except during the addition of aluminium. The aluminate solution was rapidly cooled to 50° C and filtered as quickly as possible by suction through a Schott-Jena filter 17 G4 in order to remove the insoluble black residue. The filter was previously boiled in sodium hydroxide solution, thouroughly washed with boiled distilled water and dried. The perfectly clear filtrate was diluted to 500 ml. The analysis showed on the average 1.463 *M* Al_2O_3/l. and 2.201 *M* Na_2O/l.

The experiments were performed with aluminate solutions obtained by diluting this stock-solution with water and methanol to a concentration of approximately 1 *M* Al_2O_3/l.

We began to take measurements immediately after the preparation of the aluminate solution of selected concentration, with addition of methanol or otherwise, and after cooling to +1° C. The taking of measurements was continued up to the moment of beginning precipitation. The time elapsed from the moment we began to dissolve the aluminium to the moment we began to take measurements was about one hour. The aluminate solutions were contained in stoppered Erlenmeyer flasks and automatically shaken during the whole experiment. The temperature of +1 ± 0.1° C was kept up by a water thermostat. The water in the thermostat was violently agitated and ice was constantly added.

The Al_2O_3 and Na_2O were determined by titration with hydrochloric acid with phenolphthalein and tropaeolin 00 as indicators (Hermann[10]). The recults obtained by titrating to a transition colour from onion-red to pink were in very good agreement with those obtained gravimetrically.

At fixed time-intervals samples were withdrawn from the flasks and the viscosity and specific gravity determined with an Ostwald viscosimeter and an areometer respectively, at a temperature of +1 ± 0.1° C. These determinations took about 10 minutes; the samples were then returned into the flasks. The error of viscosity measurements amounted in our experiments to less than ±1%.

RESULTS AND DISCUSSION

a) *The change of viscosity of pure aluminate solutions preceding the spontaneous precipitation of aluminium hydrate.*

These experiments were performed on three aluminate solutions of analogous composition as shown in Table I.

Curves *a* in Fig. 1 show the changes od viscosity of these aluminate solutions. The viscosity is expressed in centipoises (cP). Curves *b* show the changes of specific gravity of the same solutions.

It is evident from Fig. 1 that both viscosity and specific gravity of clear aluminate solutions undergo certain changes from the beginning of their preparation up to the incipient precipitation of aluminium hydrate. The values for viscosity and specific gravity rise and fall periodically until they reach a maximum, and then drop suddenly, this drop coinciding in time with the

TABLE I.

Composition of aluminate solutions

Solution	M Al_2O_3/l.	M Na_2O/l.	α	Spec. gravity
I	0.916	1.427	1.556	1.1890
II	0.982	1.491	1.519	1.1905
III	1.029	1.487	1.445	1.2033

precipitation of aluminium hydrate. In our experiments this occurred after about 12 to 14 hours. The two properties under investigation changed concurrently.

The differences between the lowest initial viscosity and the highest one are not large: the initial viscosities were 5.21 cP, 5.40 cP and 5.83 cP respectively, for solutions I, II and III, the viscosities at their maxima were 5.38 cP, 5.57 cP and 5.99 cP respectively. The difference between the measured time of flow of the most viscous and the least viscous of the solutions through the viscosimeter was 0.8 seconds.

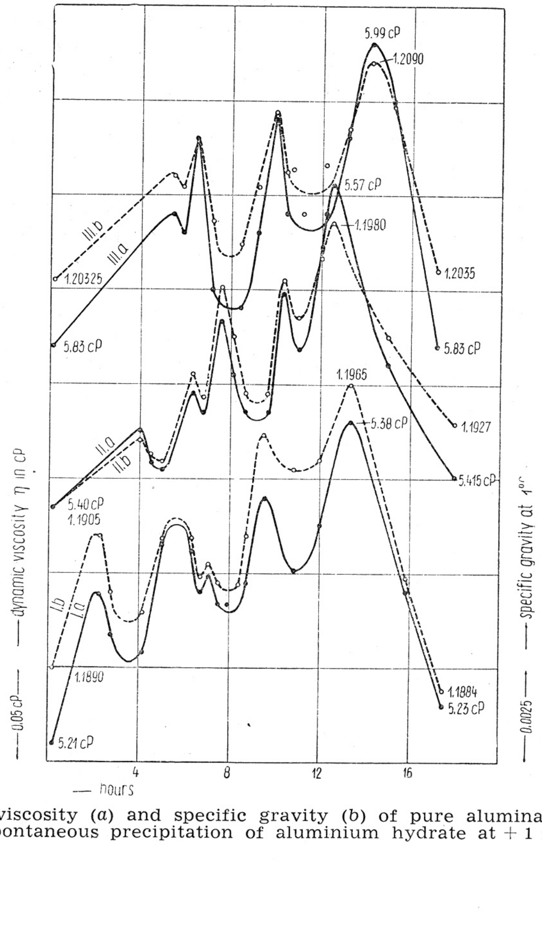

Fig. 1. Changes of viscosity (a) and specific gravity (b) of pure aluminate solutions prior to the spontaneous precipitation of aluminium hydrate at + 1 ± 0.1°C

The specific gravity changed from 1.1890 to 1.1965, from 1.1905 to 1.1980 and from 1.2033 to 1.2090 respectively for the three above mentioned solutions. The differences between the initial and the highest values are thus seen to be 0.0075 for solutions I and II, and 0.0057 for solution III.

Each point represents the mean of 5 measurements. The differences between the single measurements on the same solution were within the limits of experimental error. The phenomenon in question is thus established beyond doubt.

The increase of the values for two properties under investigation shows obviously that the structure of aluminate solutions undergoes a change prior to the precipitation of the aluminium hydrate, the size of the dissolved particles increases, probably by formation of polyaluminates: This result is in agreement with the conclusions of Iveković and Bačić,[11] which are that on dilution of unstable aluminate solutions, and just before the formation of a new phase, the following processes take place in the clear solution:

$$2\,\mathrm{Al(OH)_4^-} \longrightarrow \mathrm{Al_2(OH)_7^-} + \mathrm{OH^-} \text{ or e. g.}$$
$$2\,\mathrm{Al_2(OH)_7^-} \longrightarrow \mathrm{Al_4(OH)^-_{13}} + \mathrm{OH^-} \qquad (2)$$

and so on until in a final stage this reaction prevails:

$$\mathrm{Al_n(OH)^-_{3n+1}} \longrightarrow \mathrm{Al_n(OH)_{3n}} + \mathrm{OH^-}$$

The results point to the conclusion that in the clear solution only a relatively small proportion of the particles present is involved in this polymerisation, while the overwhelming majority of them remains in the form of meta-aluminate up to the very moment of incipient precipitation.

The curves in Fig. 1 show five maxima (with aluminate solution III the first maximum was not recorded, since there were no measurements taken during a period of six hours). This periodical change of properties may be

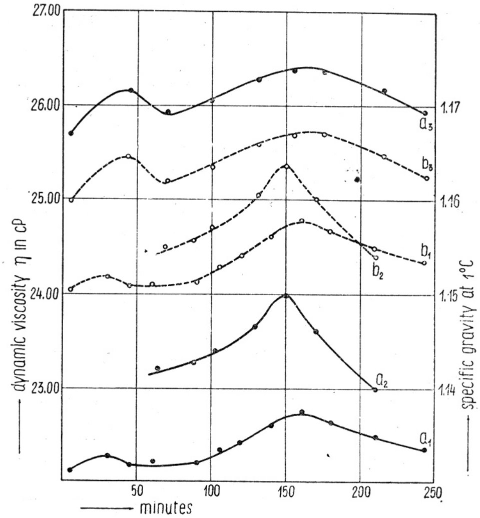

Fig. 2. Changes of viscosity (*a*) and specific gravity (*b*) of methanol-containing aluminate solutions at + 1 ± 0.1 C.

explained by the assumption that the formation of higher polyaluminates is preceded by a periodical breaking-up of the lower ones into their constituents, which are afterwards polymerized anew.

b) *The change of viscosity of aluminate solutions preceding the precipitation of aluminium hydrate in presence of methanol.*

These experiments were conducted on aluminate solutions having the same concentration of Al_2O_3 and Na_2O as those refered to in section a); to every 100 ml. of the stock-solution a determined quantity of methanol was added from a buret, and then (also from a buret) a determined quantity of water. Fig. 2 shows the changes of viscosity (*a*) and of specific gravity (*b*) of these methanol-containing aluminate solutions.

In these experiments, as in those with pure aluminate solutions, the oscillations of the properties under consideration are evident and a maximum is reached, preceding the precipitation of aluminium hydrate; in this case, however, the oscillations are not so pronounced. The initial viscosities, 22.12 cP, 23.27 cP and 25.70 cP respectively differ from the maximum viscosities — 22.76 cP, 24.00 cP and 26.46 cP respectively — by 0.64 cP, 0.73 cP and 0.66 cP respectively, i. e. by 2.9 percent on an average, which constitutes in this case a difference of full 3 seconds in the time of flow of the solution through the viscosimeter.

The specific gravities, in this case as in the foregoing, change concurrently with the viscosity.

A comparison of the initial viscosities of our methanol-containing aluminate solutions (22.12 cP, 23.27 cP and 25.70 cP respectively) with the initial viscosities of the pure aluminate solutions containing the same quantities of Al_2O_3 and Na_2O (5.21 cP, 5.40 cP and 5.83 cP respectively), or with the viscosities of pure methanol at $+1^0C$ (0.790 cP) and of pure water at $+1^0C$ (1.73 cP), shows a striking increase in viscosity caused by addition of methanol to aluminate solutions, as compared with the increase caused by the same quantity of water, despite the fact that methanol is less viscous than water. In order to find an explanation for this phenomenon, the influence of methanol on the viscosity of water, sodium hydroxide solutions and aluminate solutions of comparable concentration at $+1^0C$ was investigated.

c) *Viscosity of aqueous methanol solutions*

Mixtures of methanol and redistilled water in different proportions were prepared and their viscosities measured at $+1 \pm 0.1^0C$. The results are plotted in Fig. 3, showing the change of viscosity (η_1 in cP) as a function of concentration (in molal percent). Curve *a* shows measured viscosities at $+1 \pm 0.1^0C$, curve *a'* is a plot of Arrhenius' equation, applicable to ideal physical solutions only:

$$log \eta_s = m_{1x} log \eta_1 + m_{2x} log \eta_2 + \ldots m_{nx} log \eta_n \qquad (3)$$

where m_1, m_2 etc. are the weight rations of the components ($m_1 + m_2 + \ldots m_n = 1$), η_s the dynamic viscosity of the mixture and η_1, η_2 etc. the viscosities of the components.

The measured viscosities show a conspicuous maximum. At 1^0C the maximum is at 2.76 cP and 25.0 mole percent, which is in very close agreement with the findings of Tammann and Pillsburg[12].

The maximum may be explained by assuming a strong hydration of methanol in these mixtures. On this assumption, one molecule of methanol would bind three molecules of water at +1°C, somewhat less at higher temperatures.

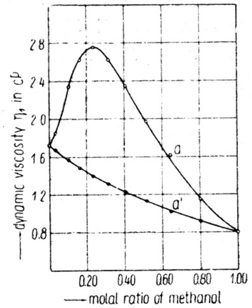

Fig. 3. Changes of viscosity of aqueous solutions of methanol at +1±0.1°C. Curve *a*- experimental values, curve *a'*- calculated values.

The difference between the measured and computed viscosity curves shows that these mixtures do not behave as ideal physical solutions and indicates that certain changes have occured in them.

d) *The viscosity of sodium hydroxide solutions and aluminate solutions in presence of methanol.*

Curve *a* in Fig. 4 is a plot of measured viscosities, curve *a'* a plot of values calculated by aid of the Arrhenius' equation analogous to the expression (3), for mixtures of methanol with an aqueous solution of sodium hydroxide. The concentration of methanol only was varied, while the concentration of sodium hydroxide was kept constant at 3.134 *M* NaOH/l., corresponding to 1.567 *M* Na_2O per litre of solution. The temperature was the same as in the foregoing experiments, i. e. + 1 ± 0.1°C.

Curves *b* and *b'* show in an analogous way the change of viscosity of mixtures of methanol with aluminate solutions as a function of methanol

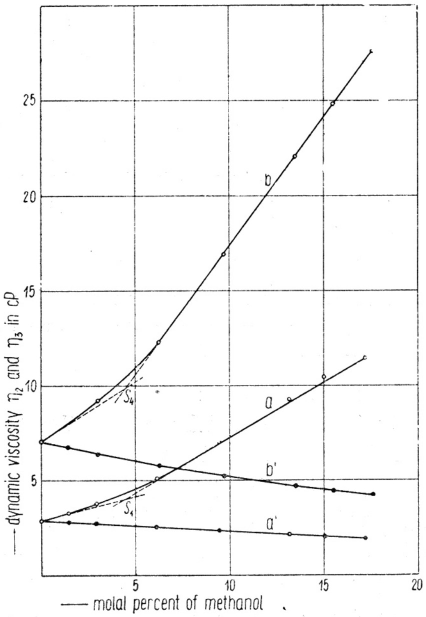

Fig. 4. Changes of viscosity of methanol-containing sodium hydroxide solutions (*a*) and aluminate solutions (*b*) at +1 ± 0.1° C. The hydroxide solutions contain 1.567 *M* Na_2O/l., the aluminate solutions 1.567 *M* Na_2O/l. and 1.029 *M* Al_2O_3/l. Curves *a'* and *b'* are calculated from equation (3).

concentration. The concentration of Al_2O_3 and Na_2O in the mixture was held constant at 1.029 M/1. and 1.567 M/1. respectively, $\alpha = 1.521$.

In order to bring into prominence the influence of the addition of methanol on the viscosity of aluminate and sodium hydroxide solutions, as compared with the analogous influence on water, the ratios η_2/η_1 and η_3/η_1 were calculated, η_2 and η_3 standing for the viscosities of methanol-containing sodium hydroxide and aluminate solutions respectively (from Fig. 4), η_1 for the viscosities of pure aqueous solutions of methanol (from Fig. 3), at equal concentrations of methanol.

In Fig. 5 the values of these ratios are plotted against the concentration of methanol (in mole percent).

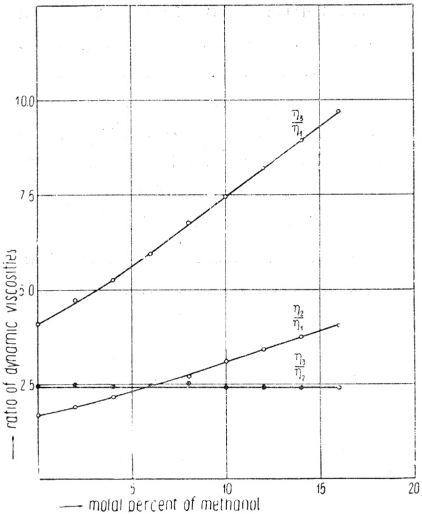

Fig. 5. Rations of the viscosities of sodium hydroxide solutions, aluminate solutions and aqueous solutions of methanol at the same concentrations of Na_2O (1.567 moles/l.) and Al_2O_3 (1.1029 moles/l.). Temperature +1 ± 0.1° C.

In the same diagram (Fig. 5) we have plotted also the values of η_3/η_2, the ratio of the viscosities of methanol-containing sodium hydroxide and aluminate solutions carrying equal concentrations of methanol, Na_2O and Al_2O_3.

Aluminate solutions with Na_2O and Al_2O_3 concentrations under consideration and more than 16 mole percent of methanol are so unstable as to render the measurements impossible. Therefore, the ratios η_2/η_1, η_3/η_1 and η_3/η_2 have been calculated for the first part of the curve in Fig. 3 only.

In what proportion the viscosity of the mixtures increases when the concentration of methanol is raised from 0 to 16 mole percent is shown by the ratio $\eta_{i(16)}/\eta_{i(0)}$ ($i = 1, 2, 3$). Thus we obtain: $\eta_{1(16)}/\eta_{1(0)} = 1{,}51$, $\eta_{2(16)}/\eta_{2(0)} = 3.68$ and $\eta_{3(16)}/\eta_{3(0)} = 3.61$.

By raising the concentration of methanol from 0 to 16 mole percent the viscosity of pure water increases 1.51 times the initial value, the viscosities of sodium hydroxide solutions 3.68 times and these of aluminate solutions 3,61 times the initial values.

The viscosities of sodium hydroxide and aluminate solutions are seen to increase in the same proportion by addition of methanol. The fact that the plot of η_3/η_2 against concentration of methanol is parallel to the axis of abscissae, shows this increase to be equal for those two solutions in the whole range of concentrations of methanol from 0 to 16 mole percent.

From $\eta_{2(16)}/\eta_{2(0)} = \eta_{3(16)}/\eta_{3(0)}$ follows

$$\eta_{3(0)}/\eta_{2(0)} = \eta_{3(16)}/\eta_{2(16)} = 2.41$$

By addition of 0 to 16 mole percent methanol the viscosity of the sodium hydroxide and aluminate solutions under consideration (of concentration 1.567 M Na_2O/1. and 1.029 M Al_2O_3/1.) increases from 2.88 to 10.6 cP for the sodium hydroxide solution and from 7.05 to 25.4 cP for the aluminate solution.

The viscosities of pure aqueous methanol mixtures are less affected by addition of methanol. They increase from 1.73 to 2.62 cP only. This points to the fact that the addition of methanol to sodium hydroxide and aluminate solutions causes the formation of larger particles in the later cases than in the former one. The fact that increasing quantities of methanol affect in the same way the viscosity of both sodium hydroxide and aluminate solutions (η_3/η_2 = const.) seems to warrant the conclusion that in both these solutions fundamentally the same process takes place.

In Fig. 6 curves *a* and *b* are plots of measured values for the dynamic viscosities of mixtures of methanol with sodium hydroxide solutions of different concentrations, while curves *a*' and *b*' are plots of the same values calculated from the Arrhenius' equation according to the expression (3).

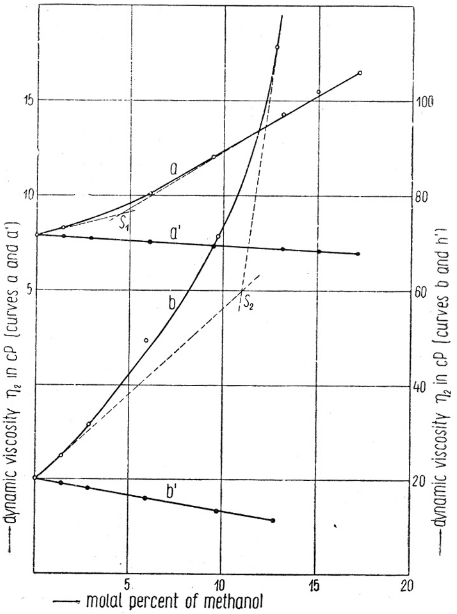

Fig. 6. Changes of viscosity of sodium hydroxide solutions of diferent concentrations. Curve *a* — 1.567 M Na_2O/l., curve *b* — 4.152 M Na_2/l. Curves *a*′ and *b*′ are calculated from equation (3). Temperature +1 ± 0.1° C.

The concentration of the sodium hydroxide solution was 3.134 M NaOH/1. (1.567 M Na_2O/1.) of mixture for curve *a,* 8.304 M NaOH per litre of mixture (4.152 M Na_2O/1.) for curve *b.*

The points of intersection S_1 at 4.5 mole percent and S_2 at 11.0 mole percent of methanol correspond to 4.54 and 10.98 mole percent of NaOH respectively. The molal ratio of methanol to sodium hydroxide is 0.993 and 1.002 respectively. This points to the fact that methanol and sodium hydroxide enter into a molecular combination according to the equation:

$$CH_3OH + NaOH \rightleftarrows CH_3OH \,.\, NaOH \qquad (4)$$

By measuring the changes of viscosity of methanol with an intermediate concentration (4.356 M NaOH/1. corresponding to 2.178 M Na_2O/1.), a point of intersection S_3 has been found at 6.1 mole percent methanol, corresponding to 6.05 mole percent NaOH. The molal ratio is 1.009 in this case.

In the face of this evidence it may be safely assumed that the molal ratio under consideration is constant in the whole range of concentrations from 3.134 to 8.304 M NaOH/1., which seems to be a strong argument in favour of our assumption of a molecular compound.

With aluminate solutions the point of intersection S_4 at 4.5 mole percent of methanol corresponds to a concentration of 2.52 moles of methanol per 1000 ml. of mixture, or 0.872 moles of methanol per mole of sodium hydroxide. The molal ratio of methanol to sodium hydroxide is thus seen to be somewhat less than one in this case. However, this may be due to the fact that a calulated value, rather than an experimentally determined one, was used for the weight concentration of sodium hydroxide in the aluminate solution. But even so it is significant that the molal ratio is approximately constant in this complex system.

Under the assumption that aluminium is present in the pure aluminate solution as the meta-aluminate ion $Al(OH)_4^-$ only, the quantity of free Na_2O per litre of solution should be 1.567—1.029 = 0.538 M, corresponding to 1.076 M free NaOH. On the other hand, if the point of intersection S_4 — in analogy to the points S_1 and S_2 — corresponds to the formation of a molecular combination, a quantity of NaOH equivalent to the methanol present, i. e. 2.52 moles, are bound up in this combination, and the difference only would be available for the meta-aluminate, i. e. 3.134—2.52 = 0.614 moles of NaOH. Since the quantity of aluminium present is 2.058 moles, which is considerably more, the conclusion is warranted that the methanol-containing aluminate solution contains polyaluminates in considerable concentration.

Assuming that only one kind of aluminate ions is present, the ratio Al/Na would be 2.058/0.634 = 3.25 what corresponds roughly to the trialuminate hydroxo-complex ion $Al_3(OH)^-_{10}$. In conjunction with the conclusions of Iveković and Bačić, this assumption seems very probable. It is supported, moreover, by the very large increase of viscosity of aluminate solutions on addition of methanol, and by the increased instability of these solutions.

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IZVOD

O promjeni viskoziteta aluminatnih lužina prije izlučivanja aluminijeva hidrata

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Da se vidi, da li se i kako se mijenja struktura aluminatne lužine u bistroj otopini prije izlučivanja aluminijeva hidrata, izvedena su mjerenja viskoziteta i spec. težine tih otopina počevši od časa priprave pa do početka izlučivanja aluminijeva hidrata.

Pokusi su rađeni s čistim aluminatnim lužinama i s aluminatnim lužinama uz dodatak metanola, koje su sadržavale 0.916 do 1.029 *M* Al_2O_3/l i 1.427 do 1.567 *M* Na_2O/l, pri čemu je kaust. modul iznosio 1.445 do 1.556.

Nađeno je, da viskoziteti čistih aluminatnih lužina, kao i aluminatnih lužina, gdje je prisutan metanol, periodički rastu i padaju sve dok ne postignu maksimum. U tom času počinje izlučivanje aluminijeva hidrata, a viskozitet otopine naglo pada.

Nađeno je, da iste koncentracije metanola u znatnijoj mjeri povećavaju viskozitet otopini natrijeva hidroksida i aluminatne lužine nego vodi. Budući da iste koncentracije metanola povećavaju u istoj mjeri viskozitet upotrebljene lužine, i natrijeve kao i aluminatne, možemo reći, da se u obje ove otopine odvijaju u osnovi isti procesi.

Nađeno je, da metanol stvara s natrijevim hidroksidom u vodenim otopinama različitih koncentracija labilni molekulski spoj molarnog odnosa 1 : 1. Taj odnos ostaje isti i u otopinama aluminatne lužine.

Jednostavnim je računom dokazano, da u takvoj metanolnoj aluminatnoj lužini postoje polialuminati u znatnijoj koncentraciji. Ako pretpostavljamo samo jednu vrstu čestica, tada je to kompleksni trialuminat hidrokso ion $Al_3(OH)^-_{10}$.

ZAVOD ZA ANORGANSKU, ANALITIČKU
I FIZIKALNU KEMIJU FARMACEUTSKOG
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